The Self Avoiding Walk as a Critical Phenomenon

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Abstract

The self avoiding walk is a simple model for the shape of a polymer chain, which naturally cannot self-intersect. Of particular interest are the set of scaling exponents that govern the shape of the chain, including the mean squared displacement between two ends of the chain. Theoretical progress was made on understanding these exponents by self-consistent field theory [1], and soon thereafter by application of renormalization group methods. It turns out that the $n \to 0$ limit of the statistical physics $O(n)$ model at criticality is mappable onto the self avoiding walk [2], and this mapping yields estimates of scaling exponents that can be checked by experiment. Here we will discuss this mapping and the insights it yields.

1 Introduction

1.1 Motivation

Our goal is to understand the shape of a single long polymer chain composed of $N \gg 1$ monomers of length a linked together. The simplest model of this system is known as the "ideal chain" and does not take into account excluded volume effect. In this model, if one end of the chain is at the origin, then the other end is at a position that is the sum of N small displacement vectors corresponding to each of the monomers. For large N this gives us a gaussian distribution of the chain end, with standard deviation $\sim \sqrt{N}a$. A natural extension of this model is to consider a situation in which the chain interacts with itself. To be concrete, we define the energy of a chain $\mathbf{r}(s)$ (parameterized by a dimensionless coordinate s which counts links along the chain) as

$$\mathcal{H}(\mathbf{r}) = \frac{1}{2}\sum_{s,t=1}^{N} U(|\mathbf{r}(s) - \mathbf{r}(t)|), \tag{1}$$

Here $U(r)$ is an interaction potential which gives us the interaction energy of two links of the chain a distance r apart.

We are interested in properties of the Gibbs measure over this Hamiltonian, where the probability of a particular chain configuration is $p(\mathbf{r}) = \frac{1}{Z}e^{-\beta\mathcal{H}(\mathbf{r})}$ for a partition function Z. For the rest of these notes we will consider $U(r) = \delta(r)$ and $\beta \to \infty$, in which case the analysis simplifies. However it seems that for short range $U(r)$ and $\beta < \infty$ the scaling results we will derive below still apply. We can proceed by asking a whole host of questions about this model whose answers we know for the ideal chain. For example, we can ask how $\langle|\mathbf{r}(N) - \mathbf{r}(0)|\rangle$ scales with N, where the average is taken over this Gibbs measure. In the case of the ideal chain this quantity scales as $N^{1/2}$ due to the central limit theorem. It is natural to expect that the scaling exponent for the self avoiding walk will be larger than $1/2$. But how much larger will it be? We define a scaling exponent ν such that $\langle|\mathbf{r}(N) - \mathbf{r}(0)|\rangle \sim N^{\nu}$, and will discuss this exponent in more detail later.

It is worth emphasizing the role of spatial dimensionality, which we shall denote as d. As in all statistical physics models, the universal results that we will discuss depend strongly on d, though not on aspects of the model such as lattice structure. We can already see the importance of d if we consider the self avoiding walk (SAW) in $d = 1$. Here trivially one finds that $\nu = 1$ for the SAW, whereas $\nu = 1/2$ for the random walk (ideal chain). As we increase d, we can see that the behavior of the SAW looks more similar to that of the ideal chain, which has $\nu = 1/2$ for all d. The general intuition is that the larger d is, the more similar these two models will be, since there is more space for the polymer to explore and therefore lower chance that it intersects itself.

2 The Self Avoiding Walk

As said previously, we will focus on the case where the walk is strictly self avoiding, corresponding to $U(r) = \delta(r)$ and $\beta \to \infty$. Our model is defined on a lattice, but approaches a continuum description as the lattice size gets very large. Before proceeding, we define some quantities with fundamental importance to characterizing this model. $c_N(\mathbf{r})$ is the the number of possible SAWs of length N, connecting the origin to point $\mathbf{r}$. We can also define $c_N = \sum_{\mathbf{r}} c_N(\mathbf{r})$ as the total number of SAWs of length N. This is conjectured to scale as

$$c_N \sim \mu^N N^{\gamma-1} \text{ for } \gamma > 1. \tag{2}$$

As one might expect $\mu < z$ where z is the coordination number of the lattice. (For an ideal chain behavior, $c_N = z^N$.) Less expected is the presence of the power law enhancement to the scaling. For now we have defined the exponent of this scaling as $\gamma - 1$ in anticipation of things to come.

2.1 Numerics

How does one numerically generate a self avoiding walk of length N? This simple question has a rather complicated answer, since it turns out that sampling a SAW of length N uniformly at random from the set of all non self-intersecting paths is quite tricky. Generating a SAW by keeping track of visited sites and avoiding them tends to favor paths which stray less far from the origin, as discussed in Amit, Parisi and Peliti (1983) [3]. In fact the upper critical dimension for this model — the "true" self avoiding walk or tSAW — is $d = 2$, since above this dimension the self intersection of a random walk becomes a sub-leading correction. One way to see this is that the number of intersections scales as $N^2/N^{d/2}$ and this term becomes sublinear and therefore a subdominant contribution to R_N^2 for $d > 2$. The tSAW and the SAW define different probability measures over the set of non-intersecting paths of length N. For the tSAW, the measure defined is equivalent to that defined by a simple random walk in $d > 2$.

In lieu of the tSAW model for generating self-avoiding paths, there are many Monte Carlo algorithms in both the canonical (fixed N) and grand canonical (variable N) ensembles that can be used to sample SAWs from the appropriate uniform measure. An in-depth review is given in Sokal 1994 [4]. Here we use the algorithm described in Berretti and Sokal 1985 [5], which produces SAWs with an exponential distribution of lengths with a variable average determined by parameter choices. We simply use this algorithm for visualization purposes here, but care is needed to make quantitative conclusions from these numerics.

2.2 Flory Argument

A simple and surprisingly accurate estimate of the exponent ν is due to Paul Flory. Flory's argument comes from balancing two contributions to the free energy of the polymer chain. The first is the repulsive energy: the smaller a volume the chain is crammed into, the larger the repulsive potential energy it has. This contribution to the free energy tends to make the chain swell into a larger volume. The second is the entropy: there are more ways to organize the chain with small end-to-end distance than with large end-to-end distance. This contribution causes the chain to be more compact.

We can estimate the repulsive energy of the chain by saying that is is proportional to the concentration of monomers squared, since they have to interact with one another, integrated over the volume of the chain. If the radius of the volume occupied by the chain is R, the number of monomers is N and the spatial dimension is d, this gives us

$$E_{\text{repulsive}} \sim \epsilon \frac{N^2}{R^d}, \tag{3}$$

where ϵ is a dimensional constant quantifying the energy penalty of the chain interacting with itself.

To estimate the entropy of the polymer coiled into a shape of linear dimension R, we note that the number of ways to have this shape is proportional to the probability density that the polymer has this size. Assuming that the size of the polymer, r, is gaussian distributed with $p(r) \sim e^{-r^2/Na^2}$, where a is monomer length, we find that the entropic contribution of a configuration of size R is

$$E_{\text{entropic}} = S_0 - \frac{R^2}{Na^2}. \tag{4}$$

Putting together these two contributions tells us that

$$E_{\text{total}} \sim \epsilon \frac{N^2}{R^d} + T\frac{R^2}{Na^2}. \tag{5}$$

This expression is minimized for $N^2 R^{-d-1} \sim R/N$. We can therefore deduce that

$$R \sim N^{\frac{3}{d+2}}. \tag{6}$$

This estimate that $\nu = \frac{3}{d+2}$ is incredibly accurate, matching experiments much better than can be expected. Note that in our estimate of the entropy we used the expression for an ideal chain. This significantly overestimates the entropy, since the denominator of the entropy terms should scale superlinearly in N for a self repelling chain. However we also significantly overestimated the repulsive energy of the chain since we did not account for correlations in the positions of adjacent monomers, instead assuming they are independently distributed inside a volume R^d. It turns out that these two errors cancel each other out to a remarkable degree, but there doesn't seem to be a clear way to know this ahead of time.

2.3 Upper Critical Dimension

We have seen that the Flory argument gives $\nu = \frac{3}{d+2}$ for spatial dimension d. This implies that $\nu = 1/2$ for $d = 4$. This is simply the ideal chain exponent! In addition the Flory argument gives $\nu < 1/2$ in $d > 4$, but we know this must be nonsense. It turns out that in fact for $d > 4$ the exponents of the SAW are identical to those of the ideal chain, which performs a simple random walk. This already gives us a hint that there may be some similarity with the $O(n)$ model, in which the upper critical dimension is also 4.

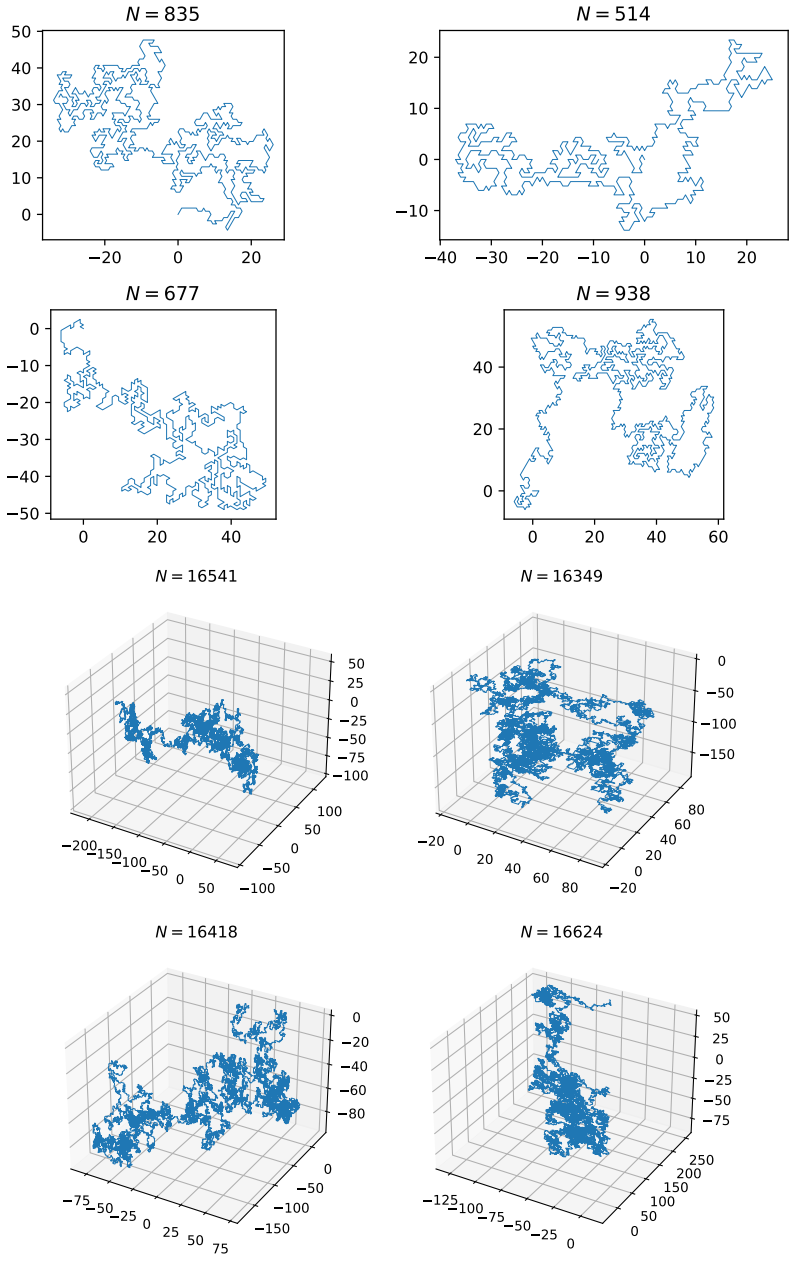

Figure 1: The self avoiding walk in 2 and 3 dimensions, for various runs of the Monte Carlo algorithm terminated after some large number of steps.

We can see more explicitly why the upper critical dimension is 4 with a scaling argument. We know that R should scale at least as fast as $N^{1/2}$; therefore we can upper bound the repulsive energy by

$$E_{\text{repulsive}} \lesssim \frac{N^2}{N^{d/2}}. \tag{7}$$

Similarly we can lower bound the entropic energy by

$$E_{\text{entropic}} \gtrsim \frac{(\sqrt{N})^2}{Na^2} = \mathcal{O}(1). \tag{8}$$

Therefore we see that the repulsive interactions become irrelevant for large N when $2 - d/2 < 0$, i.e. $d > 4$.

2.4 Self Consistent Approach

A more principled approach to this problem was established by Edwards [1]. This approach consisted of setting up a self consistent formulation of the problem where the polymer performs a walk in a potential, which is itself generated by the walk. This is similar to the Hartree-Fock approximation for calculating a many body wavefunction in the presence of a potential created by by the particles described by the wavefunction itself. The results of this rather involved calculation, amazingly, give the same answer as the Flory estimate in $d = 2, 3, 4$. In order to do better than this, there seem to be two main approaches. The first is to expand the Greens function for a self repelling polymer with small parameter v_0 which represents the excluded volume. This method had been pursued prior to de Gennes, and gets quite labor intensive to compute terms in the expansion. However de Gennes notices a correspondence between this approach and a statistical physics model of a magnet in a particular limit [2]. It is this correspondence that we pursue and attempt to elucidate here.

3 The $O(n)$ Model

Here we take a slight detour and define the model of a magnet that ends up being useful to understand the SAW. We model each spin as an n-component vector with length normalized to $\sqrt{n}$, and place spins at the lattice sites in d dimensions. We denote the components of a spin at spatial location $\mathbf{r}$ as $S_\alpha(\mathbf{r})$, where α indexes the component. Normalization enforces that $\sum_\alpha S_\alpha(\mathbf{r})^2 = n$ (this normalization will be important when we take $n \to 0$). The Hamiltonian of the system is given by

$$\mathcal{H} = -K \sum_{\langle ij \rangle} \mathbf{S}(\mathbf{r}_i) \cdot \mathbf{S}(\mathbf{r}_j), \tag{9}$$

where the $\{\mathbf{r}_i\}$ are restricted to lattice sites, and the sum is over nearest neighbor bonds which tend to align the spins. We can see that that $n = 1$ case of this model is exactly the Ising model, $n = 2$ is exactly the XY model, and $n = 3$ is the classical Heisenberg model. One can therefore write down the partition function of this system as

$$Z = \int e^{-\mathcal{H}/\tau} \prod_i d\mathbf{S}_i, \tag{10}$$

where the integral is over all the degrees of freedom of all the spins, which correspond to the angular degrees of freedom of each spin (since the magnitudes are fixed).

4 Mapping the SAW to the $O(n)$ Model

4.1 Calculating a correlation function

Our goal is now to write an expression for the correlation function of spins in the $O(n)$ model that might be useful for understanding the SAW. The correlation function of interest is

$$\langle S_\alpha(0) S_\alpha(\mathbf{r}) \rangle = \frac{1}{Z} \int S_\alpha(0) S_\alpha(\mathbf{r}) e^{-\mathcal{H}/\tau} \prod_i d\mathbf{S}_i. \tag{11}$$

Note that α is the component index of interest: there is no summation over α even though it is repeated. We also abbreviate notation for this correlation function as $\langle S_{0\alpha} S_{r\alpha} \rangle$

We will rewrite the exponential in the integrand here as if we were doing a high-temperature expansion of the correlation function, but it will turn out that this is exact in the limit $n \to 0$. We expand the exponential in the integrand only to first order in K/τ, and suppress the higher order terms.

$$\langle S_{0\alpha} S_{r\alpha} \rangle = \frac{1}{Z} \int S_{0\alpha} S_{r\alpha} \prod_{\langle ij \rangle} \exp\left(\frac{K}{\tau} \mathbf{S}_i \cdot \mathbf{S}_j\right) \prod_i d\mathbf{S}_i \approx \int S_{0\alpha} S_{r\alpha} \prod_{\langle ij \rangle} \left(1 + \frac{K}{\tau} \mathbf{S}_i \cdot \mathbf{S}_j\right) \prod_i d\mathbf{S}_i, \tag{12}$$

where the notation $\prod_{\langle ij \rangle}$ indicates a product over nearest neighbor sites i and j.

What happens when we expand out all the terms in this product? In total, if the number of bonds in the lattice is L, there will be 2^L terms in the expansion, where each term is comprised of a subset of the bonds, multiplied by the appropriate power of K/τ, which is the number of bonds in the subset.

When the integral is performed, many such terms vanish by symmetry. In fact, all terms which do not have two factors of each spin component vanish. As an example, consider one spatial dimension where $r = 2$. The only term from our correlation function that can contribute, once the integral is performed, is

$$\left(\frac{K}{\tau}\right)^2 S_{0\alpha} (\mathbf{S}_0 \cdot \mathbf{S}_1)(\mathbf{S}_1 \cdot \mathbf{S}_2) S_{2\alpha}. \tag{13}$$

This immediately tells us that the terms containing components of $\mathbf{S}$ other than α must vanish, and so the relevant contribution is even simpler: $(K/\tau)^2 S_{0\alpha}^2 S_{1\alpha}^2 S_{2\alpha}^2$. Under the integral, this simply contributes $(K/\tau)^2$.

Therefore the terms which remain correspond to subsets of bonds which form a continuous path between the origin and $\mathbf{r}$. However (in $d > 1$) this path can have loops and self intersections and all manner of contortions. Furthermore, so far we have dealt only with the first order expansion of the exponential in the integrand for the correlation function. What about higher order terms? For the same symmetry reasons these will only contribute if they are comprised of spin products which form a continuous path from the origin to $\mathbf{r}$, but this will still complicate things.

It turns out that in one fell swoop, we can eliminate both 1) the paths between 0 and $\mathbf{r}$ that intersect themselves and 2) the contributions from the higher order terms in the expansion! *We do this by setting* $n = 0$.

[Side note: if one wants to be careful, then one should also calculate Z itself, which involves worrying about closed loops (including loops between adjacent spins, to which the quadratic terms in the expansion of $e^{-\mathcal{H}/\tau}$ can contribute. However when we set $n = 0$ the contribution of all the loops vanishes and we get $Z = 1$.]

4.2 Setting $n = 0$ eliminates unwanted terms

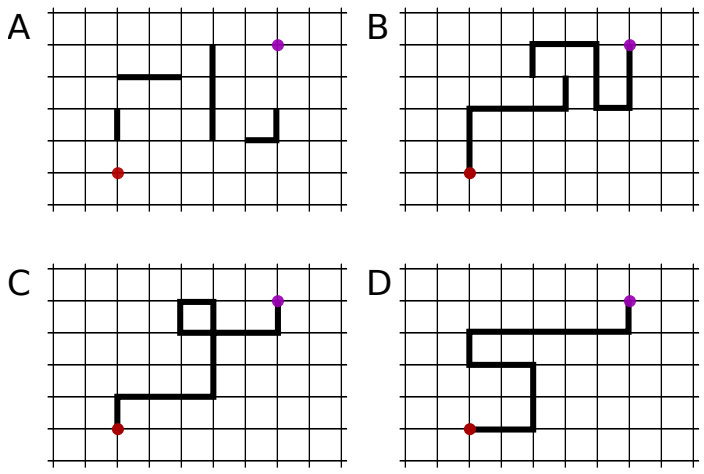

Figure 2: A schematic of the terms that contribute to the correlation function between spins at the origin (in red) and $\mathbf{r}$ (in purple). The terms in A) and B) vanish due to the contributions of unpaired spins, and so when the average is performed, flipping the unpaired spin will cancel out the contribution of the term. These terms would vanish even without the $n \to 0$ limit. The term in C) vanishes as $n \to 0$ due to the fact that there is a spin that participates in this term 4 times, and any terms of order higher than 2 must vanish when $n \to 0$ as we show below. Therefore the only terms that contribute are those of the type shown in D): self-avoiding paths from the origin to $\mathbf{r}$. The contribution of such a term is $(K/\tau)^N$ where N is the length of the path.

Here we show why this seemingly unphysical limit generates a correlation function of the self avoiding walk. Consider the generating function (or Laplace transform) of a single spin vector $\mathbf{S}$, given by

$$f(\mathbf{k}) = \int d\mathbf{\Omega} e^{-\mathbf{k}\cdot\mathbf{S}}. \tag{14}$$

This function allows us to calculate any moment of any combination of the components of this spin, by taking derivatives and evaluating at $k = 0$. Namely,

$$\int d\mathbf{\Omega} S_{\alpha_1} \cdots S_{\alpha_\ell} = (-1)^\ell \frac{\partial}{\partial_{k_{\alpha_1}}} \cdots \frac{\partial}{\partial_{k_{\alpha_\ell}}} f(\mathbf{k}) \Big|_{k=0}. \tag{15}$$

By symmetry $f(\mathbf{k})$ can only depend on the magnitude of $\mathbf{k}$, and so we can write it as $f(k)$ without any loss of information.

We can calculate

$$\sum_\alpha \frac{\partial^2}{\partial k_\alpha^2} f = \int d\mathbf{\Omega} \sum_\alpha S_\alpha^2 e^{-\mathbf{k}\cdot\mathbf{S}} = nf. \tag{16}$$

Furthermore since $\partial k/\partial k_\alpha = k_\alpha/k$, we can calculate the same expression in a different way; namely

$$\sum_\alpha \frac{\partial^2}{\partial k_\alpha^2} f = \frac{\partial^2 f}{\partial k^2} + \left(\frac{n-1}{k}\right) \frac{\partial f}{\partial k}. \tag{17}$$

Therefore setting these equal gives

$$\frac{\partial^2 f}{\partial k^2} + \left(\frac{n-1}{k}\right) \frac{\partial f}{\partial k} - nf = 0. \tag{18}$$

Now when we set $n = 0$ we are left with the equation $f''(k) - k^{-1} f'(k) = 0$. (Here the reason for the normalization choice $\sum_\alpha S_\alpha^2 = n$ becomes apparent.) The appropriate boundary conditions are $f(0) = 1$ and $f'(0) = 0$, which come from the definition of f along with symmetry considerations. Therefore the appropriate solution of the equation for f (when $n = 0$) is $f(k) = 1 - k^2/2$. This tells us that all moments higher than second order in any component of a spin will vanish. Furthermore, all first moments are also vanishing.

4.3 Putting it all together

What does this mean for our correlation function? If we set $n = 0$, then we only need to worry about the the continuous non self-intersecting paths between the origin and $\mathbf{r}$. We have shown this via three important points.

1. The only terms that are quadratic in the $\mathbf{S}$ components that can contribute to the correlation function $\langle S_{0\alpha} S_{r\alpha} \rangle$ correspond to paths with endpoints at the origin and $\mathbf{r}$.
2. Setting $n = 0$ in the $O(n)$ model eliminates contributions from all of these paths which cross themselves.
3. Setting $n = 0$ also eliminates contributions from terms which are higher than quadratic in the $\mathbf{S}$ components.

Each path from 0 to $\mathbf{r}$ of length N contributes $(K/\tau)^N$ to the correlation function when the integral is performed. Therefore we have established the following rather amazing relation:

$$\langle S_\alpha(0) S_\alpha(\mathbf{r}) \rangle|_{n=0} = \sum_N c_N(\mathbf{r}) \left(\frac{K}{\tau}\right)^N. \tag{19}$$

This is the fundamental correspondence that will allow us to transfer understanding of critical phenomena to understanding of the SAW.

4.4 Analogies between SAW and $O(n = 0)$

If we are to claim a similarity between the $O(n)$ model at criticality and the self avoiding walk, there is a mismatch that we must address. The self avoiding walk is not, at least not apparently, a system which is critical: it does not sit at the boundary of a spontaneous symmetry-broken phase, and there does not seem to be a parameter analogous to temperature in the $O(n)$ model, which we can tune to change the distance from criticality.

In fact we can show that in a precise sense, the SAW is critical and that parameter which tunes degree of criticality is the length of the walk, N. We can see this first heuristically by recasting the scaling relationships of the SAW in terms of known relationships for the $O(n)$ magnet. For the magnet we know that the correlation length ξ is related to the reduced temperature $t = \frac{\tau - \tau_c}{\tau_c}$ by a power law divergence as $\tau_r \to 0$, characterized by the exponent ν according to

$\xi \sim |t|^{-\nu}$. We know that for the self avoiding walk, the end-to-end separation obeys a scaling with N that is given by $R \sim N^{\nu} = (1/N)^{-\nu}$. Therefore as we send $1/N \to 0$, we have a similar type of scaling relationship to the more familiar one from the magnetic system. In this sense it is productive to think of $1/N$ as an analog of the reduced temperature for the SAW.

To formalize this mapping, recall the conjecture that

$$c_N \sim \mu^N N^{\gamma-1}. \tag{20}$$

We can show that this is consistent with the susceptibility of the $O(n=0)$ magnet diverging as $\chi \sim |t|^{-\gamma}$ at the critical temperature. This also justifies our definition of γ in the SAW model to match γ as commonly defined in the study of critical phenomena. Recall that the susceptibility, given by $\partial_H \langle M \rangle$ where $\langle M \rangle = -\tau \partial_H \log Z$, can be written in terms of the two point correlation function as

$$\chi_\alpha = \int d^d\mathbf{r} \langle S_\alpha(0) S_\alpha(\mathbf{r}) \rangle. \tag{21}$$

Translating this into the corresponding expression from the SAW and putting in the conjectured form of c_N (and dropping the subscript on χ) we have

$$\chi = \sum_N \mu^N N^{\gamma-1} \left(\frac{K}{\tau}\right)^N. \tag{22}$$

One can see that this quantity diverges when $\tau = K\mu$. If we identify τ_c with μK then $\log(\tau/\mu K) \approx \frac{\tau - \tau_c}{\tau_c} = t$ and we can write

$$\chi = \sum_N e^{-tN} N^{\gamma-1}. \tag{23}$$

Converting this sum to an integral and changing variables to $N' = tN$, one can see that χ diverges as $t^{-\gamma}$ as $t \to 0$ from above. This is the sense in which the SAW is critical. The value of μ, which depends on lattice details, corresponds to a critical temperature τ_c in the magnet model, which is also nonuniversal. However the critical exponents are universal. We also see that the reduced temperature t in the magnet model is conjugate to N, the length of the chain, in the SAW. Therefore $N \to \infty$ corresponds to $t \to 0$, and brings us closer to criticality.

4.5 The wandering exponent ν

We are now in a position to refine the Flory estimate of the wandering exponent ν, defined so that $\langle R^2 \rangle \sim N^{2\nu}$. We can guess that R is the SAW analog of the correlation length ξ from the magnetic model, based on the fact that both are lengths. In principal it should also be possible to look at divergence of the correlation length as $t \to 0$ based on the two point correlation function $\langle S_{0\alpha} S_{r\alpha} \rangle|_{n=0}$, and thereby figure out what the corresponding quantity with the same critical exponent as ξ is in the SAW. However here we will simply guess this answer, which turns out to be correct. Once we identify ξ with R, since we have the correspondence $t \sim 1/N$, we get $R \sim N^{\nu}$ where ν is the critical exponent of the correlation in the $O(n=0)$ model.

Luckily for us, there has already been a lot of work done on deriving critical exponents for the $O(n)$ model, and we can simply co-opt these results for our purposes. The upper critical dimension of the $O(n)$ model is 4, and Wilson and Fisher established a procedure of expanding around 4 dimensions in parameter $\epsilon = 4 - d$ to calculate critical exponents in dimensions lower than the upper critical dimension. To first order in ϵ, these results give us

$$\frac{1}{\nu} = 2 - \frac{n+2}{n+8}\epsilon. \tag{24}$$

Setting $\epsilon = 1$ (for $d = 3$) and $n = 0$ gives $\nu = 4/7 \approx 0.571$. This is close to $\nu = 0.6$ from Flory, but slightly smaller! And, with some effort, one can extend this approach to higher orders in ϵ. A more accurate value is $\nu \approx 0.5882$ in 3 dimensions [6]. It turns out that this rather minute difference can actually be observed in experiment! [7]

It is worth remarking that for the renormalization group (RG) calculation required to obtain ν for the $O(n)$ model, the limit $n \to 0$ has a natural interpretation. In evaluating the RG transformation one generally has to do a diagrammatic perturbation expansion of an integral over a field. In the diagrams that result, setting $n = 0$ eliminates contributions from all diagrams with loops!

5 Further Reading

Scaling Concepts in Polymer Physics by de Gennes [8] offers much of this material from the horse's mouth.
Scaling and Renormalization in Statistical Physics by Cardy [9] has a nice discussion of the mapping between the $O(n=0)$ model and the SAW.

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